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Double 1s shell ionization of Si induced in collisions with protons and heavy ions

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Abstract

The double 1s ionization of Si induced in collisions with protons and heavier ions (C, Ne) was studied by measuring the K X-ray emission of a solid Si target. In order to resolve the hypersatellite contributions in the spectra, high-resolution crystal diffractometry was employed yielding subelectronvolt energy resolution. Experimentally obtained hypersatellite yields were used to determine the double to single K shell ionization cross-section ratios σ_{KK}/σ_K corresponding to the investigated collisions. The experimental ratios for collisions with heavy ions, where direct Coulomb ionization and electron capture need both to be considered, were compared to the theoretical values calculated within the independent electron approximation employing single electron ionization probabilities calculated by the three body classical trajectory Monte-Carlo (CTMC) method. For proton collisions where direct ionization solely contributes to 1s ionization the semiclassical approximation (SCA) was employed.

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1. Introduction

High-resolution measurements of K X-ray spectra provide valuable information about the multiple inner shell ionization. In the case of double 1s ionization, photons emitted in radiative transitions correspond to hypersatellite lines in the K X-ray spectra. Different processes can lead to double 1s ionization, the highest cross-sections being observed in collisions with heavy ions. Several experiments concerning collisions of heavy ions on low Z elements have been performed in the past [1–3]. Due to the high probability for additional ionization in outer shells resulting in complex X-ray

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spectra, the hypersatellite intensities could not be determined accurately in these works. On the other hand in experiments with protons, where additional outer shell ionization is less probable, double ionization cross-sections of some low Z elements could be obtained [4]. Similar was the case with heavy ions on mid-Z atoms [5] where additional outer shell ionization was also negligible.

In the case of asymmetric collisions ($Z_{\text{projectile}} \ll Z_{\text{target}}$) the dominant ionization mechanism is the direct Coulomb ionization. For more symmetric collisions the capture of inner shell electrons into the empty shells of the projectile (electron transfer) may become very important and in some cases even prevails. A detailed investigation of the relative importance of the two ionization mechanisms has been done by Hall et al. [6,7] for Ti bombarded by heavy ions.

In the case of Si the hypersatellites are shifted above the Kβ diagram line and overlap exactly the KβL satellites. The only possibility to resolve experimentally the two overlapping groups of lines is to use high-resolution X-ray spectroscopy employing crystal spectrometers. A theoretical modeling of the target X-ray emission is needed in addition to extract properly the hypersatellite yields. Mostly due to the above-mentioned problems quantitative experimental data about double 1s shell ionization of Si induced in ion–atom collisions are extremely scarce.

2. Experiment and data analysis

The experiment with heavy ions was performed at the variable energy cyclotron of the Paul Scherrer Institute in Villigen, Switzerland. A thick Si target (0.23 g/cm^2) was bombarded by 34 MeV C^{2+} and 50 MeV Ne^{3+} ions. Beam intensities of 10–100 nA were used. The target K X-ray emission spectra (Fig. 1) were measured with a high-resolution von Hamos Bragg crystal spectrometer presented in detail in [8]. First order reflection on the $(1\bar{1}0)$ SiO_2 crystal was used. For the energy calibration Kα X-ray spectra of Mg, Al, Si and P induced by the bremsstrahlung from an X-ray tube were used.

Since at the fixed crystal-detector position the energy acceptance of the spectrometer was about

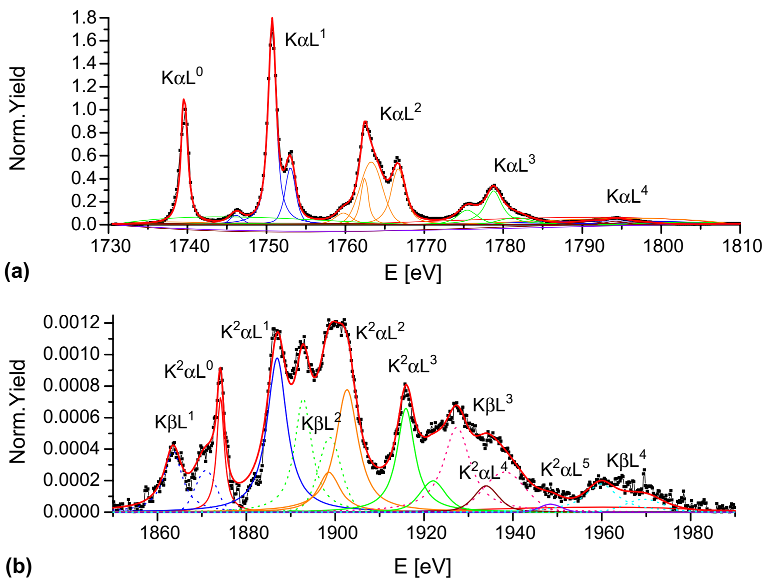

Fig. 1. Measured satellite (a) and hypersatellite (b) X-ray spectra of Si produced by impact of 34 MeV C ions. The spectra were fitted with pseudo-Voigt curves describing the MCDF calculated lineshapes. Kα (1s $\rightarrow$ 2p) contributions are shown by solid lines, Kβ (1s $\rightarrow$ 3p) by dotted lines.

40 eV, the total K X-ray spectrum had to be measured in seven separate parts. An overlap of about 100 channels was chosen in order to normalize the intensity of the neighboring spectra. Due to the inhomogeneous beam intensity profile on the target, the normalization was not straightforward leading to final uncertainties of 20–30% for the intensities of the seventh spectrum. At the end the measured intensities were corrected to account for the detector efficiency and the crystal reflectivity.

Experiment with heavy ions has been extended towards asymmetric collisions with protons. Measurements with 2 MeV protons were performed at the J. Stefan Institute, Ljubljana employing the 2 MV tandem accelerator and crystal spectrometer in Johansson geometry. Beam currents of 1–2 μA were used. Photons emitted from the target, which was positioned well inside the Rowland circle at a distance of 37 cm in front of the crystal, were diffracted in first order by the $(1\bar{1}0)$ reflecting planes of a SiO_2 crystal. The radius of the Rowland circle was 50 cm. Since the measured $K\alpha^hL^0$ and $K\beta L^1$ lines practically coincide in energy, their relative intensities are affected by neither the energy dependent detector efficiency nor the crystal reflectivity.

3. Results and discussion

3.1. Asymmetric collisions with heavy ions

The Kα satellite and hypersatellite photon yields were extracted by fitting the spectra using the appropriate number of pseudo-Voigt curves corresponding to the theoretical lineshape of each contribution deduced from calculations with the GRASP92 computer code [9]. In order to determine the initial state production yields from the corresponding photon yields, decay schemes of multiply ionized Si atoms were calculated. Since measurements were performed on thick target, self-absorption correction incorporating stopping of the projectiles in the target and absorption of X-rays on their way out, was also needed to obtain the ionization cross-section ratio at the impact energy of the projectile. Details about this rather complex data analysis are given in [10]. The double to single K shell ionization cross-section ratios were determined as:

$$\frac{\sigma_d}{\sigma_s} = \frac{\sum_{N=0}^{5}\sigma(K^2L^N)}{\sum_{N=0}^{5}\sigma(KL^N)} = \frac{\sum_{N=0}^{5}I(K^2L^N)}{\sum_{N=0}^{5}I(KL^N)}. \tag{1}$$

The results are presented in Table 1. The quoted errors originate from the experimental errors, the uncertainties in the calculated decay scheme and the uncertainties related to the overlap of the Kβ satellite and Kα hypersatellite lines (approximately 15%).

The cross-section ratio was also calculated within the independent electron model whose validity has been demonstrated several times for multiple ionization induced in ion–atom collisions [22,23]. We have considered direct ionization and electron capture and used single ionization probabilities obtained within the three body classical trajectory Monte-Carlo simulation (CTMC) [11]. The used model and initialization parameters are described in detail in [12]. In the calculation only the strongest K–K transfer channel was considered. For the L shell ionization only direct ionization, which dominates, was taken into account. The average projectile charge state determined from experimental values tabulated in [13] was used in the calculations. The calculated σ_{KK}/σ_K ratios are given in Table 1. The calculated values are in agreement with the experimental ones.

3.2. Symmetric collisions with protons

For the proton measurements a slightly different approach was adopted since only the $K\alpha^hL^0$ hypersatellite contribution can be measured. As a

Table 1
Experimental and theoretical double to single K shell ionization ratios in percents for the measured collisions

	σ_{KK}/σ_K theory	Exp.
Si + 34 MeV C	11.6	9.6 ± 1.9
Si + 50 MeV Ne	9.0	9.0 ± 2.3
Si + 2 MeV p	12.3×10^{-2}	$(5.2 \pm 1.2) \times 10^{-2}$

For collisions with heavy ions the theoretical cross-section ratios were calculated using the CTMC single electron ionization/capture probabilities, while for collisions with protons SCA model was used.

consequence, only the leading term in Eq. (1) remains significant and can be written as follows:

$$\frac{\sigma_{\rm d}}{\sigma_{\rm s}} = \frac{I({\rm K}^2{\rm L}^0)}{I({\rm KL}^0)} = \frac{I({\rm K}^2{\rm L}^0)}{I({\rm KL}^1)}\frac{I({\rm KL}^1)}{I({\rm KL}^0)} \tag{2}$$

The second term in Eq. (2) can be determined separately from measurements of the Kα diagram and KαL^1 satellite lines [14], whereas the first term can be obtained from the measured Kα^hL^0 and KβL^1 spectra presented in Fig. 2. In order to obtain ionization cross-section ratio the corresponding X(Kα^2L^0) and X(KβL^1) X-ray yield ratio can be written as

$$\frac{X({\rm K}\alpha^2{\rm L}^0)}{X({\rm K}\beta{\rm L}^1)} = \frac{I({\rm K}^2{\rm L}^0)\omega^{\alpha}_{\rm KK}}{I({\rm KL}^1)(1-R)\omega^{\beta}_{\rm KL}} \Rightarrow \frac{I({\rm K}^2{\rm L}^0)}{I({\rm KL}^1)} = \frac{X({\rm K}\alpha^2{\rm L}^0)}{X({\rm K}\beta{\rm L}^1)}(1-R)\frac{\omega^{\beta}_{\rm KL}}{\omega^{\alpha}_{\rm KK}} \tag{3}$$

The rearrangement factor R in case of Si equals $R = 0.109 \pm 0.033$ [14]. The Kα and Kβ partial fluorescence yield ratio for KK and KL ionized atoms has been calculated using total K shell fluorescence yields as calculated by Tunnel and Bhalla [15] multiplying them by the $\Gamma_{K\beta}/\Gamma^{\rm rad}_{\rm K} \approx \Gamma_{K\beta}/\Gamma_{K\alpha}$ value given by Scofield [16]. Additional factor of 5/6 was considered in $\Gamma^{\rm rad}_{\rm K}$ due to the reduced number of 2p electrons available for the Kα transition according to Larkins [17].

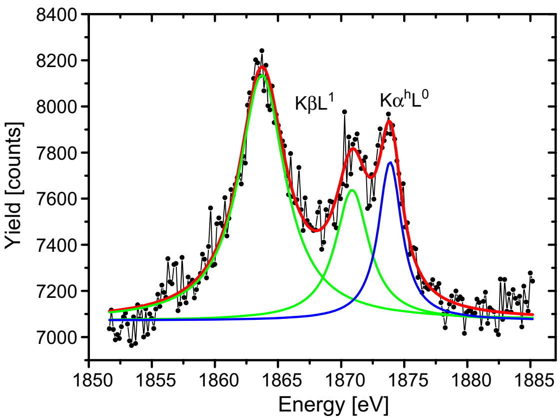

Fig. 2. Measured Kα^hL^0, KβL^1 spectrum of Si produced by impact of 2 MeV protons. The hypersatellite line was fitted with a single Lorentzian. Excellent agreement of the energy of the fitted line (1873.9 ± 0.2 eV) with the recent calculation by Martins et al. [21] yielding 1874 eV for Si, confirm that the modeled Lorentzian correspond to the Kα^hL^0 hypersatellite transition. The spectrum can be compared to the corresponding part of the C-induced spectrum (Fig 1(b)). The Kα^hL^0/KβL^1 intensity ratio is much smaller in the proton-induced spectrum indicating a lower $\sigma_{\rm KK}/\sigma_{\rm K}$ cross-section ratio.

Finally the measured KβL^1 intensity needs to be corrected for the shake contribution, while for the 1s^2 state the shake contribution can be neglected [18]. The shake contribution of the KβL^1 intensity was estimated by comparing the recently measured KαL^1 shake satellite intensity [19] with the KαL^1 satellite intensity induced by 2 MeV protons that we have measured recently [14]. The intensity ratio obtained from this comparison was adopted also for the KβL^1 satellite line, since the Kβ and Kα satellites represents just different decays of the same initial state.

The double to single 1s direct ionization cross-section ratio obtained in this way was compared to the theoretical value calculated within the semi-classical approximation (SCA) using the first order SCA model of Trautmann and Rösel (IONHYD code) [20]. Binding energies of the united atom model were employed and multipoles up to the order $l = 5$ were considered in the calculations. As shown in Table 1, the SCA value overestimates significantly the experimental one.

4. Summary and conclusion

The K X-ray spectra of Si induced in collisions with heavy ions and protons were measured by means of high-resolution X-ray spectroscopy, using curved crystal spectrometers. The main objective of the study was to determine the double to single 1s ionization cross-section ratios related to the different collisions. Thanks to the subelectronvolt energy resolution of the employed spectrometers, we were able to extract the yields of the Kα hypersatellite which overlaps the KβL satellite line. From the experimentally determined initial hypersatellite production yields the $\sigma_{\rm KK}/\sigma_{\rm K}$ ionization cross-section ratios were determined for the measured collisions. Relatively high cross-section ratios of about 10% were obtained for the collisions with C and Ne ions, an approximately 200 times smaller value being found for the 2 MeV proton collision.

Whereas for asymmetric collisions with protons the 1s shell ionization is governed by the direct Coulomb ionization, the electron capture becomes significant for more symmetric collisions involving heavy ions. It was shown that for the asymmetric proton–silicon collisions the measured σ_{KK}/σ_K ratio is overestimated by the SCA model whereas for heavy ion collisions CTMC calculations incorporating both ionization mechanisms give values that are in relatively good agreement with the experimental results.

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